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(54) Novel reaction product and ORI-inhibited motor fuel composition.

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(57) A novel gasoline-soluble reaction product, a concentrate comprising the reaction product dissolved in a hydrocarbon solvent, and a haze-free, deposit resistant and ORI-inhibited motor fuel composition comprising the reaction product is described. The reaction product is obtained by reacting a dibasic acid anhydride, a novel diamine containing block copolymers with polyoxyalkylene backbones, and a hydrocarbyl polyamine. Motor fuel compositions containing the reaction product additive of the instant invention are haze-free and show improved deposit-resistance and ORI control in comparison with motor fuels without the reaction product. Motor fuel compositions of the instant invention may optionally comprise a polyolefin polymer/copolymer component having a molecular weight range of about 500-3500.

NOVEL REACTION PRODUCT AND ORI-INHIBITED MOTOR FUEL COMPOSITION

BACKGROUND OF THE INVENTION

Field of the Invention

5 This invention relates to a gasoline-soluble reaction product, to a concentrate comprising the reaction product dissolved in a hydrocarbon solvent, and to a motor fuel composition comprising the reaction product. More particularly, this invention relates to: (i) the reaction product of a dibasic acid anhydride, a novel diamine containing block copolymers with polyoxyalkylene backbones, and a hydrocarbyl polyamine;
 10 (ii) a concentrate comprising the reaction product dissolved in a hydrocarbon solvent; and (iii) a motor fuel composition comprising the reaction product.

Combustion of a hydrocarbonaceous motor fuel in an internal combustion engine generally results in the formation and accumulation of carbon deposits on various parts of the combustion chamber as well as on the fuel intake and exhaust systems of the engine. Any accumulation of deposits in the combustion chamber seriously reduces the operating efficiency of the engine. First, deposit accumulation within the combustion chamber inhibits heat transfer between the chamber and the engine cooling system. This leads to higher temperatures within the combustion chamber, resulting in increases in the end gas temperature of the incoming charge. Consequently, end gas auto-ignition occurs, which causes engine knock. Second, the accumulation of deposits within the combustion chamber reduces the volume of the combustion zone,
 20 causing a higher than design compression ratio in the engine. This, in turn, also results in serious engine knocking. A knocking engine does not effectively utilize the energy of combustion. Moreover, a prolonged period of engine knocking will cause stress fatigue and wear in vital parts of the engine. The above-described phenomenon is characteristic of gasoline powered internal combustion engines. It is usually overcome by employing a higher octane gasoline for powering the engine, and hence, has become known as the engine "octane requirement increase" (ORI) phenomenon. It would therefore be highly advantageous if engine ORI could be substantially reduced or eliminated by preventing deposit formation in the combustion chamber of the engine.

An additional problem common to internal combustion engines relates to the accumulation of deposits in the carburetor which tend to restrict the flow of air through the carburetor at idle and at low speed, resulting in an overrich fuel mixture. This condition also promotes incomplete fuel combustion and leads to rough engine idling and engine stalling. Excessive hydrocarbon and carbon monoxide exhaust emissions are also produced under these conditions. It would therefore be desirable from the standpoint of engine operability and overall air quality to provide a motor fuel composition which minimizes or overcomes the above-described problems.

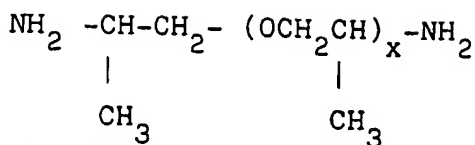
35 Deposit-inhibiting additives for use in motor fuel compositions are well known in the art. However, conventional additives may cause hazing of the motor fuel. Hazy motor fuels are unacceptable by the public since they may indicate a problem with the fuel, such as the presence of undesired contaminants. It would therefore be desirable to provide a haze-free motor fuel composition which is deposit-resistant and ORI-inhibited.

40 The present invention provides a gasoline-soluble reaction product composition which, when employed in a motor fuel composition, reduces the tendency of the motor fuel to form deposits. Consequently, by employing in motor fuel compositions the reaction product of the present invention, engine ORI is substantially reduced. Furthermore, use of the present reaction product can provide motor fuel compositions which are haze-free, clear, and stable.

2. Background Art

Our US-A-4,581,040 (Sung et al.) teaches the use of a reaction product as a deposit inhibitor additive in fuel compositions. The reaction product taught is a condensate product of the process comprising:

(i) reacting a dibasic acid anhydride with a polyoxyisopropylenediamine of the formula

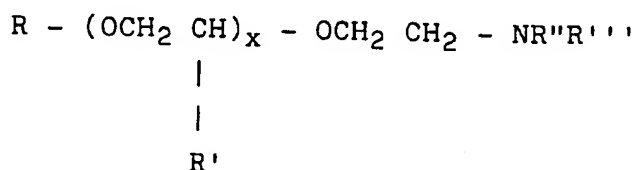


where x is a numeral of about 2-50, thereby forming a maleamic acid;

(ii) reacting said maleamic acid with a polyalkylene polyamine, thereby forming a condensate product; and

(iii) recovering said condensate product.

US-A-4,604,103 (Campbell) discloses a motor fuel deposit control additive for use in internal combustion engines which maintains cleanliness of the engine intake system without contributing to combustion chamber deposits or engine octane requirement increase (ORI). The additive disclosed is a hydrocarbyl polyoxyalkylene polyamine ethane of molecular weight range 300-2500 having the formula

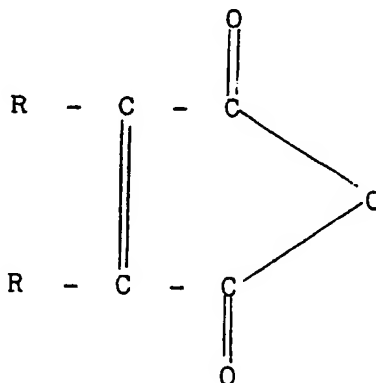


where R is a hydrocarbyl radical of from 1 to about 30 carbon atoms; R' is selected from methyl and ethyl; x is an integer from 5 to 30; and R'' and R' are independently selected from hydrogen and $(\text{CH}_2\text{CH}_2\text{NH})_y\text{H}$ where y is an integer from 0-5.

SUMMARY OF THE INVENTION

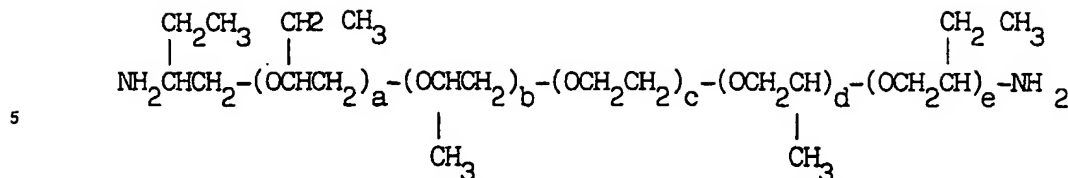
It has been discovered that a composition comprising the reaction product of (a) a dibasic acid anhydride, (b) a diamine containing block copolymers with polyalkylene backbones, and (c) a hydrocarbyl polyamine, has utility as an ORI inhibitor when employed as a soluble additive in a motor fuel composition. The novel reaction product of the instant invention is obtained by reacting at a temperature of 30°C-200°C, preferably 90°C-150°C:

(a) about 1 mole of a dibasic acid anhydride of the formula



Where R₁ is either H or C₁-C₅ alkyl radical, preferably H;

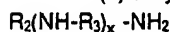
(b) 1-2 moles, preferably 1.5 moles, of a polyoxyalkylene diamine of the formula



where c has a value of from 5 to 150, preferably 8-50; b + d has a value of from 5 to 150, preferably 8-50; and a + e has a value of from 2 to 12, preferably 4-8; and

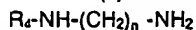
(c) 1-2 moles, preferably 1 mole, of a hydrocarbyl polyamine which may be either:

(1) a hydrocarbyl polyamine of the formula



where R_2 is a alkyl radical having from 1 to 24, preferably 12 to 20, carbon atoms, R_3 is an alkylene radical having from 1 to 6 carbon atoms, and x has a value from 1 to 10, preferably 1 to 5; or

(ii) a n-alkyl-alkylene diamine of the formula



where R_4 is an aliphatic hydrocarbon radical having from 1 to 24, preferably 12 to 20 carbon atoms and n has a value of from 1 to 6, preferably 3.

The instant invention also provides a concentrate comprising from 1.0 to 75.0 weight percent, preferably 5.0-35.0 weight percent, of the prescribed reaction product dissolved in a hydrocarbon solvent, preferably xylene. In addition, the instant invention provides a haze-free motor fuel composition comprising from 0.0005 to 5.0, preferably 0.001-1.0, most preferably 0.01-0.1, weight percent of the prescribed reaction product. An additional polymer/copolymer additive with a molecular weight range from 500 to 3500, preferably 650-2600, may also be employed in admixture with the motor fuel composition of the instant invention in concentrations of from 0.001 to 1.0 wt. %, preferably 0.01-0.5 wt. %.

BRIEF DESCRIPTION OF THE DRAWINGS

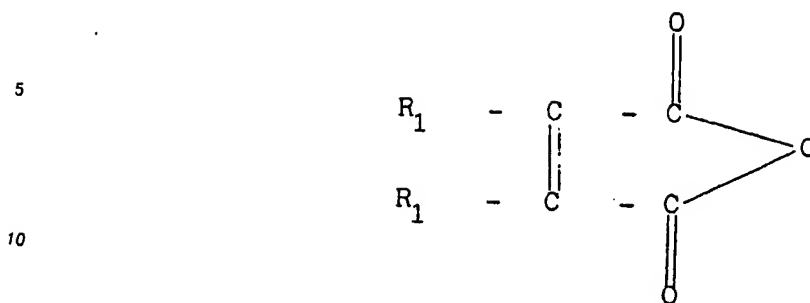
Referring now to the drawings, Figure 1 is a graphical representation of data obtained which compares the octane requirement (as a function of hours of engine operation) of a 1983 Chevrolet 2.0 liter engine using an unleaded base fuel containing 60 PTB of a commercial fuel additive, and the identical engine using a motor fuel composition of the instant invention which is an unleaded base fuel containing 100 PTB of the reaction product of the instant invention, as exemplified by Example II.

Similarly, Figure 2 is a graphical representation of data obtained which compares the octane requirement (as a function of hours of engine operation) of a 1083 Chevrolet 2.0 liter engine using an unleaded base fuel containing 60 PTB of a commercial fuel additive, and the identical engine using a motor fuel composition of the instant invention, as exemplified by Example VI, which is an unleaded base fuel containing 30 PTB of the reaction product of the instant invention, as exemplified by Example II, in combustion with 150 PTB of polyisobutylene of a molecular weight of about 1300.

DETAILED DESCRIPTION OF EMBODIMENTS OF THE INVENTION

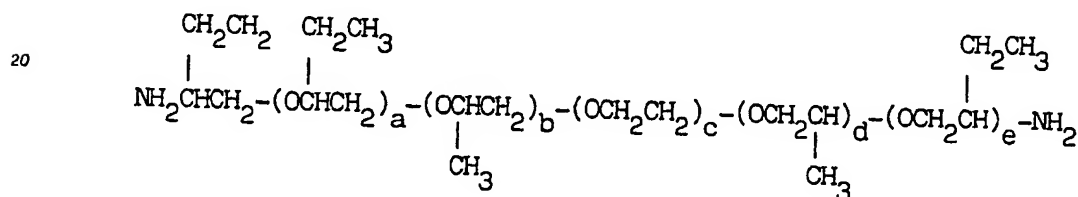
The novel reaction product of the instant invention is prepared by reacting a dibasic acid anhydride, a diamine containing block copolymers with polyoxyalkylene backbones and a hydrocarbyl polyamine.

The dibasic acid anhydride reactant used to prepare the reaction product is of the formula



where R₁ is either H or a C₁-C₅ alkyl radical. Accordingly, dibasic acid anhydrides suitable for use include maleic anhydride; alpha-methyl maleic anhydride; alpha-ethyl maleic anhydride; and alpha, beta-dimethyl maleic anhydride. The preferred dibasic acid anhydride for use in maleic anhydride.

The novel polyoxyalkylene diamine reactant used to prepare the reaction product is a diamine of the formula



where c has a value of from about 5 to about 150, preferably 8-50; b + d has a value of from about 5 to about 150, preferably 8-50; and a + e has a value of from about 2 to about 12, preferably 4-8. A distinctive feature of the prescribed polyoxyalkylene diamine reactant resides in the fact that it contains a large number (5-150, preferably 8-50) of polyoxypropylene ether moieties in combination with a smaller number (2-12, preferably 4-8) of polyoxybutylene ether moieties. The best mode of synthesizing the prescribed polyoxyalkylene diamine reactant is set forth in Example I, below.

Example I

Synthesis of Novel Polyoxyalkylene Diamine Reactant

A. Preparation of Polyol Precursor

4.54 kg (ten pounds) of a polyethylene glycol of an approximate molecular weight of 600 and 100 g of 45% aqueous KOH were charged into a 38 x 10³ m (ten-gallon) reactor, which was then purged with prepurified nitrogen. While maintaining a nitrogen purge, the reactor was heated to 100°C, and the initiator was then dried to a water content of less than 0.1% by vacuum stripping followed by nitrogen stripping. Thereafter, 8.67 kg (19.1 lb) of ethylene oxide was charged and reacted at 105-110°C and a gauge pressure of 345 x 10³ pa (50 psig) for 1.25 hours. Without digestion 11.89kg (26.2 lb) of propylene oxide was then charged and reacted at 105-110°C and a gauge pressure of 345 x 10³ Pa (50 psig) over a 3 hour period.

The reaction mixture was thereafter heated to about 120°C, and 1.32 kg (2.9 lb) of butylene oxide was added over a 30 minute period. After a 2 hour digestion period, the alkaline polyol was neutralized by stirring for 2 hours with 360. g of absorbive magnesium silicate (MAGNESOL 30/40), which was added as an aqueous slurry. To stabilize the material, 26.4 g of di-t-butyl p-cresol was added. The neutralized product was then vacuum stripped to a pressure of about 6.67 x 10² Pa (5 mm Hg), nitrogen stripped, and filtered. The finished product had the following properties:

Acid no., mg KOH/g	0.01
Hydroxyl no., mg KOH/g	35
Water, wt %	0.01
pH in 10:6 isopropanol-water	8.1
Color, Pt-Co	40

	Sodium, ppm	0.2
	Potassium, ppm	0.2
	Peroxide, ppm	1.1
	Viscosity, °F, cc	
5	77	988
	100	513

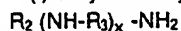
B. Amination Reaction

0.27 kg/hr (0.6 lb/hr) of the polyol, 0.54 kg/hr 1.2 lb/hr) of ammonia, and 36 liter/hr of hydrogen were fed into a 1270 ml tubular reactor filled with a nickel-chromium-copper metal and metal oxide catalyst which was kept at 200°C and 13.8×10^6 Pa (2000 psig). The reactor effluent was stripped at 100°C and (10mm Hg vacuum). The resulting polyether polyamine product had the following physical properties.

	Total acetylatables, meq/g	0.615
	Total amaine, meq/g	0.56
	Primary amine, meq/g	0.54
	Water, wt %	0.09
20	Color, Pt-Co	30
	Flash Point, pmcc	227°C (440°F)
	Melting point, °C	27-31

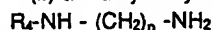
The hydrocarbyl polyamine reactant used to prepare the reaction product may be either:

(i) a hydrocarbyl polyamine of the formula



Where R_2 is an alkyl radical having from 1 to about 24, preferably 12-30 carbon atoms, R_3 is an alkylene radical having from 1 to about 6 carbon atoms, and x has a value from 1 to 10, preferably 1-5; or

(ii) a n-alkyl-alkylene diamine of the formula



where R_4 is an aliphatic hydrocarbon radical having from 1 to about 24 carbon atoms, preferably from 12 to 20 carbon atoms, and n has a value from 1 to about 6, preferably having a value of 3. N-alkyl-alkylene diamines suitable for use in preparing the reaction product of the instant invention include aliphatic diamines commercially available from Akzo Chemie America Co. under the DUOMEEN series trade name. Examples of such n-alkyl-alkylene diamines include n-coco-1,3 diamino-propane (DUOMEEN C), n-soya-1,3-diaminopropane (DUOMEEN S), n-tallow-1,3-diaminopropane (DUOMEEN T), and n-oleyl-1,3-diaminopropane (DUOMEEN OL). The most preferred n-alkyl-alkylene diamine reactant for use in preparing the reaction product is n-tallow-1,3 diaminopropane.

The reaction product is prepared by first reacting about 1 mole of dibasic acid anhydride with about 1 to 2 moles, preferably 1.5 moles, of the prescribed novel diamine containing block copolymers with polyoxyethylene, polyoxypropylene and polyoxybutylene backbones at a temperature of from 30°C to 200°C, preferably 90°C-150°C. The reaction of dibasic acid anhydride with the novel polyoxyalkylene diamine is preferably carried out in the presence of a solvent. A preferred solvent is one which will distill with water azeotropically. Suitable solvents include hydrocarbons boiling in the gasoline boiling range of about 30°C to about 200°C. Generally, this will include saturated and unsaturated hydrocarbons having from about 5 to about 10 carbon atoms. Specific suitable hydrocarbon solvents include hexane, cyclohexane, benzene, toluene, and mixtures thereof. Xylene is the preferred solvent. The solvent can be present in an amount of up to about 90% by weight of the total reaction mixture. The reaction mixture is thereafter cooled to a temperature of from 50°C to 75°C, preferably 60°C, and 102 moles, preferably 1 mole, of the hydrocarbyl polyamine is added. The new mixture is then reacted at 30°C - 200°C, preferably 90°C-150°C.

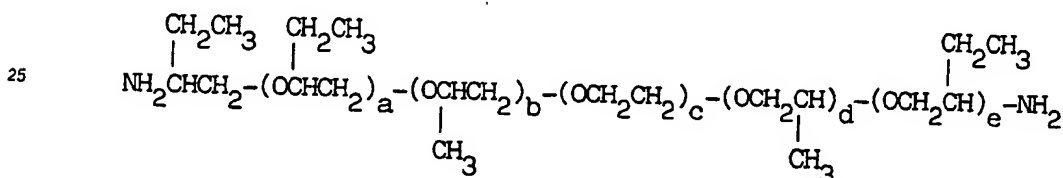
In the best mode of preparing the reaction product of the instant invention, about 1 mole of maleic anhydride and about 1.5 moles of the prescribed polyoxyalkylene diamine, where c has a value of 8-50, b+d has a value of 8-50, and a+e has a value of 4-8, are combined with the solvent xylene and reacted at a temperature of about 100°C. The reaction mixture is maintained at this temperature for approximately 2 hours. The mixture is then cooled to about 60°C, whereupon about 1 mole of the hydrocarbyl polyamine n-tallow-1, 3 diaminopropane is added. The new mixture is then reacted at about 140°C for reflux and azeotropeing for 5 hours, with from about 1 to about 1.5 moles of water being removed. The reaction product can then be separated from the solvent using conventional means, or left in admixture with some or all of the solvent.

A critical feature of the reaction product composition of the instant invention is the presence of a large number (5-150, preferably 8-50) of polyoxypropylene ether moieties in combination with more limited numbers (2-12, preferably 4-8) of polyoxybutylene ether moieties. These moieties are provided by the prescribed novel polyoxyalkylene diamine reactant. In particular, the presence of a large number of polyoxypropylene ether moieties enhances the gasoline solubility of the reaction product, thus increasing the efficacy of the reaction product as an additive in motor fuel compositions. The reaction product additive of the instant invention is advantageous over other ORI-controlling motor fuel additives in that the reaction product of the instant invention is soluble in gasoline and similar motor fuel compositions, and therefore requires no admixing with a solvent prior to introduction into a base motor fuel composition. In addition, the presence of polyoxybutylene ether moieties in the reaction product of the instant invention has been found to prevent hazing in a motor fuel composition of the instant invention.

The following Example II, III and IV (no Example I) illustrate the preferred method of preparing the novel reaction product of the instant invention. In the Examples, all parts are parts by weight unless otherwise specified.

Example II

A reaction product was formed by reacting 54 parts of maleic anhydride, 3265 parts of xylene, and 3000 parts of a polyoxyalkylene diamine at 100°C for 2 hours. The polyoxyalkylene diamine was of the formula.

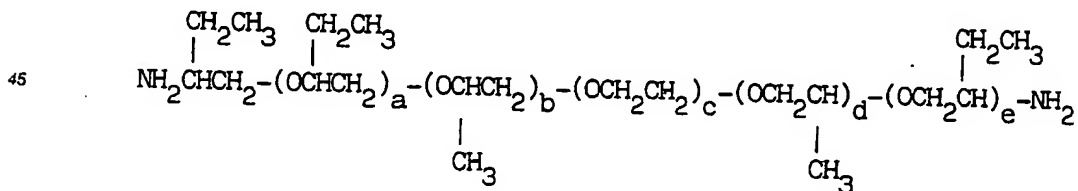


where c had an approximate value of 5-150, b+d had an approximate value of 5-150, and a+e had an approximate value of 2-12.

The mixture was therefore cooled to about 60°C, and 54 parts of n-tallow-1,3 diaminopropane (DUOMEENT) were added. The new mixture was then reacted at about 140°C for 5 hours to produce the final reaction product. The final reaction product was then filtered and stripped of remaining solvent under vacuum.

Example III

A reaction product is formed by reacting 54 parts of maleic anhydride, 3206 parts of xylene, and 3000 parts of a polyoxyalkylene diamine at 100°C for 2 hours. The polyoxyalkylene diamine is of the formula



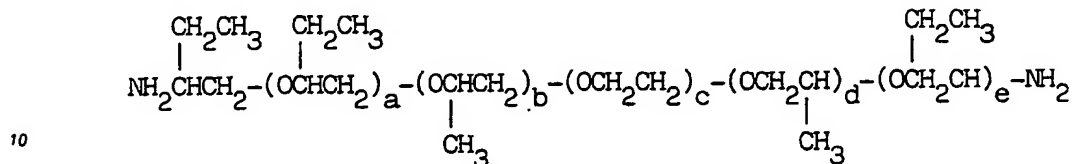
where c has an approximate value of 5-150, b+d has an approximate value of 5-150, and a+e has an approximate value of 2-12.

The mixture is thereafter cooled to about 60°C, and 152 parts of n-coco-1, 2 diaminopropane (DUOMEEN C) are added. The new mixture is then reacted at about 140°C for 5 hours to produce the final reaction product. The final reaction product is then filtered and stripped of remaining solvent under vacuum.

Example IV

A reaction product is formed by reacting 54 parts of maleic anhydride, 3231 parts of xylene, and 3000 parts of a polyoxyalkylene diamine at 100°C for 2 hours. The polyoxyalkylene diamine is of the formula.

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where c has an approximate value of 5-150, b+d has an approximate value of 5-150, and a+e has an approximate value of 2-12.

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The mixture is thereafter cooled to about 60°C, and 176 parts of n-octyl-1, 3 diaminopropane (DUOMEEN OL) are added. The new mixture is then reacted at about 140°C for 5 hours to produce the final reaction product. The final reaction product is then filtered and stripped of remaining solvent under vacuum.

20

It has been found that a motor fuel composition containing from 0.0005 to 5.0 weight percent, preferably from 0.001 to 1.0, most preferably from 0.01 to 0.1 weight percent, of the reaction product of the instant invention is surprisingly effective in minimizing and reducing the ORI of a gasoline internal combustion engine. This improvement has been demonstrated in engine tests where the performance characteristics of a base motor fuel composition containing a commercial fuel additive and an improved motor fuel composition of the instant invention were compared. The specific engine tests were made on a 2.0 liter 1983 Chevrolet four cylinder engine (Chevy Test). This test correlates well with results obtained via road simulation tests.

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The base motor fuel employed in the tests (herein designated as Base Fuel A) was a premium grade gasoline essentially unleaded (less than 0.05 g of tetraethyl lead per gallon), and comprised a mixture of hydrocarbons boiling in the gasoline boiling range and containing about 22% aromatic hydrocarbons, 11% olefinic carbons, and 67% paraffinic hydrocarbons, boiling in the range from about 32°C to about 232°C (about 90°F to about 450°F). In preparing motor fuels for the Chevy Test, a suitable amount of the reaction product of the instant invention was added directly to Base Fuel A without any hazing of the motor fuel composition, and without additional solvents being necessary. As previously stated, the gasoline solubility of the reaction product of the instant invention is attributed to the presence of a large number of polyoxypropylene ether moieties in combination with polyoxyethylene and polyoxybutylene ether moieties. The haze-free property of the motor fuel composition comprising the reaction product is attributed to the presence of the polyoxybutylene ether moieties.

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The ORI tendencies of Base Fuel A containing 60 PTB of a commercial fuel additive (60 pounds of reaction product per 1000 barrels of gasoline, equivalent to about 0.02 weight percent of reaction product based on the weight of the fuel composition), as well as Base Fuel A containing 100 PTB of the reaction product of Example II (100 pounds of reaction product per 1000 barrels of gasoline, equivalent to about 0.033 weight percent of reaction product based upon the weight of the fuel composition) were measured using the Chevy Test. The Chevy Test employs a 2.0 liter 1983 Chevrolet in-line four cylinder engine with a cast alloy iron cylinder head having separate intake and exhaust ports for each cylinder. An electronically controlled fuel injection system maintains the required fuel flow to each engine cylinder by monitoring various engine operating parameters (e.g manifold absolute pressure, throttle valve position, coolant temperature, engine r.p.m., and exhaust gas oxygen content) and adjusting the fuel flow accordingly. The fuel system supplying fuel to the engine is specifically adapted for the determination of engine ORI. At the beginning of the engine rating procedure, a fuel with an octane rating high enough to ensure that no audible engine knock is present is employed. The next lower octane fuel is then switched with the previous fuel, and this procedure continues until a knock becomes audible. The difference between the octane level at knock and no-knock conditions is the engine ORI. Engine ORI was determined as a function of hours of engine operation for both Base Fuel A containing 100 PTB of reaction product, and for Base Fuel A containing 60 PTB of a typical commercial motor fuel additive.

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The experimental results obtained from the Chevy Test for Base Fuel A containing 60 PTB of commercial fuel additive and Base Fuel A containing 100 PTB of the reaction product of the instant invention (Example II) are set forth in Figure 1. As illustrated by Figure 1, the octane requirement of the engine using Base Fuel A containing 60 PTB of commercial fuel additive was consistently higher than the

corresponding octane requirement of the engine using Base Fuel A containing 100 PTB of Example II over the duration of the test. The data set forth in Figure 1 thus indicate that the reaction product of the instant invention is more effective as an ORI controlling additive in a motor fuel composition than a typical commercially available motor fuel composition.

5 The motor fuel composition of the instant invention comprises a major amount of a base motor fuel and 0.0005-5.0 weight percent, preferably 0.001-1.0, most preferably 0.01-0.1 weight percent of the above-described reaction product. Preferred base motor fuel compositions for use with the reaction product additive are those intended for use in spark ignition internal combustion engines. Such motor fuel compositions, generally referred to as gasoline base stocks, preferably comprise admixture of hydrocarbons boiling
10 in the gasoline boiling range, preferably from about 33°C (90°F) to about 232°C (450°F). This base fuel may consist of straight chains or branched chains or paraffins, cycloparaffins, olefins, aromatic hydrocarbons, or mixtures thereof. The base fuel can be derived from, among others, straight run naphtha, polymer gasoline, natural gasoline, or from catalytically cracked or thermally cracked hydrocarbons and catalytically reformed stock. The composition and octane level of the base fuel are not critical and any conventional
15 motor fuel base can be employed in the practice of this invention. In addition, the motor fuel composition may contain any of the additives generally employed in gasoline. Thus, the fuel composition can contain anti-knock compounds such as tetraethyl lead compounds, anti-icing additives, upper cylinder lubricating oils, and the like.

The motor fuel composition of the instant invention may additionally comprise a polymeric component,
20 present in a concentration ranging from about 0.001 to about 1.0 weight percent, preferably 0.01-0.5 weight percent, based on the total weight of the motor fuel composition. The polymeric component may be a polyolefin polymer, copolymer, or corresponding hydrogenated polymer or copolymer of a C₂-C₆ unsaturated hydrocarbon. The polymer component is prepared from monoolefins and diolefins, or copolymers thereof, having an average molecular weight in the range from about 500 to about 3500, preferably from
25 about 650 to about 2600. Mixtures of olefin polymers with an average molecular weight falling within the foregoing range are also effective. In general, the olefin monomers from which the polyolefin polymer component is prepared are unsaturated C-C hydrocarbons. Specific olefins which may be employed to prepare the polyolefin polymer component include ethylene, propylene, isopropylene, butylene, isobutylene, amylene, hexylene, butadiene, and isoprene. Propylene, isopropylene, butylene, and isobutylene are
30 particularly preferred for use in preparing the polyolefin polymer component. Other polyolefins which may be employed are those prepared by cracking polyolefin polymers or copolymers of high molecular weight to a polymer in the above-noted molecular weight range. Derivatives of the noted polymers obtained by saturating the polymers by hydrogenation are also effective and their use in an optional feature of this invention. The word "polymers" is intended to include the polyolefin polymers and their corresponding
35 hydrogenated derivatives.

The average molecular weight range of the polymer component is a critical feature. The polyolefin polymer, copolymer, or corresponding hydrogenated polymer or copolymer component may have an average molecular weight in the range from about 500 to about 3500, preferably from about 650 to about 2600. The most preferred polymer components for use in the instant invention are polypropylene with an
40 average molecular weight in the range of about 750 to about 1000, preferably about 800, and polyisobutylene with an average molecular weight in the range of from about 1000 to about 1500, preferably about 1300. The polymer component, if employed, enhances the ORI reduction of the instant invention, and additionally provides enhanced cleanliness at the engine intake valves and ports.

45 Examples V and VI, set forth below, are illustrative of motor fuel compositions of the instant invention comprising the above-described reaction product and polymer components.

Example V

50 A motor fuel composition was obtained by mixing with Base Fuel A about 100 PTB of the reaction product component set forth in Example II (Equivalent to about 0.033 wt. %) and about 150 PTB of polypropylene polymer component of a molecular weight of about 800 (equivalent to about 0.05 wt. %)

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Example VI

A motor fuel composition was obtained by mixing with Base Fuel A PTB of the reaction product component set forth in Example II (equivalent to about 0.01 wt %) and about 150 PTB of polyisobutylene of a molecular weight of about 1300 (equivalent to about 0.5 wt. %).

Chevy Test data comparing ORI for Base Fuel A containing 60 PTB of commercial fuel additive and a motor fuel composition of the instant invention (Example VI), which is Base Fuel A containing 30 PTB of the reaction product of Example II combined with 150 PTB of polyisobutylene of a molecular weight of about 1300 are set forth in Figure 2. As illustrated by Figure 2, the octane requirement of the engine using Base Fuel A containing 60 PTB of commercial fuel additive was consistently higher than the corresponding octane requirement of the engine using a motor fuel composition of the instant invention over the duration of the test. The data set forth in Figure 2 thus indicate that the reaction product of the instant invention, in admixture with the prescribed optional polymer component, is more effective as an ORI controlling additive in a motor fuel composition than a typical commercially available motor fuel composition.

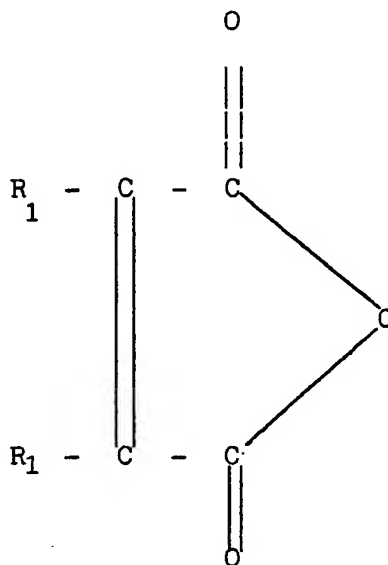
For convenience in shipping and handling, it is useful to prepare a concentrate of the reaction product additive which maybe added to a base motor fuel to produce the motor fuel composition of the instant invention. The concentrate may be prepared in a suitable liquid solvent containing from about 1.5 to about 75.0 weight percent, preferably 5.0-35.0 weight percent, of the additive component or components, that is to say, the above-described novel reaction product either alone or in combination with the above-described additional polymer component. Suitable solvents for use in the concentrate include hydrocarbon solvents such as toluene and xylene, with xylene being preferred.

The terms and expressions employed herein are used as terms of description and there is no intention, in the use of these descriptive terms and expressions, of excluding equivalents of the features described.

Claims

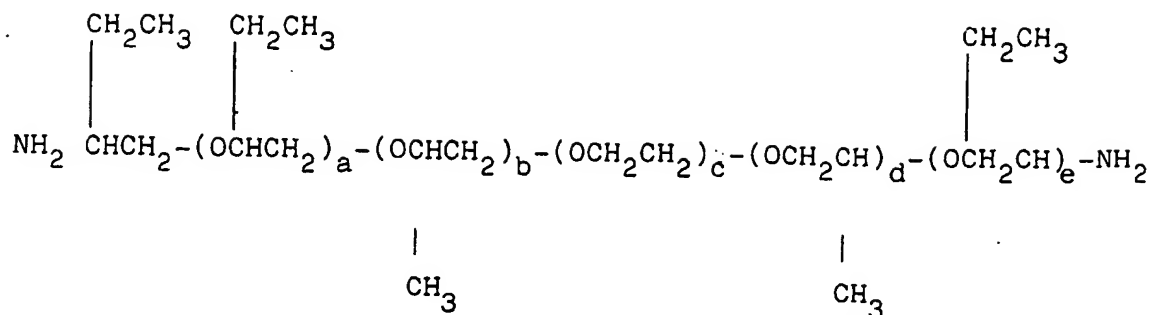
1. A composition characterised in that it comprises the reaction product obtained by reacting at a temperature in a range of from 30°C to 200°C:

(a) about 1 mole of a dibasic acid anhydride of the formula



where R₁ is either H or a C₁-C₅ alkyl radical;

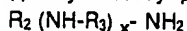
(b) 1-2 moles of a polyoxyalkylene diamine of the formula



where c has a value of from 5 to 150, b + d has a value of from 5 to 150, and a + e has a value of from 2 to 12; and

- 15 (c) 1-2 moles of a hydrocarbyl polyamine which may be either:

(i) a hydrocarbyl polyamine of the formula



where R_2 is a alkyl radical having from 1 to 24 carbon atoms, R_3 is an alkylene radical having from 1 to 6 carbon atoms, and x has a value of from 1 to 10; or

- 20 (ii) a n-alkyl-alkylene diamine of the formula

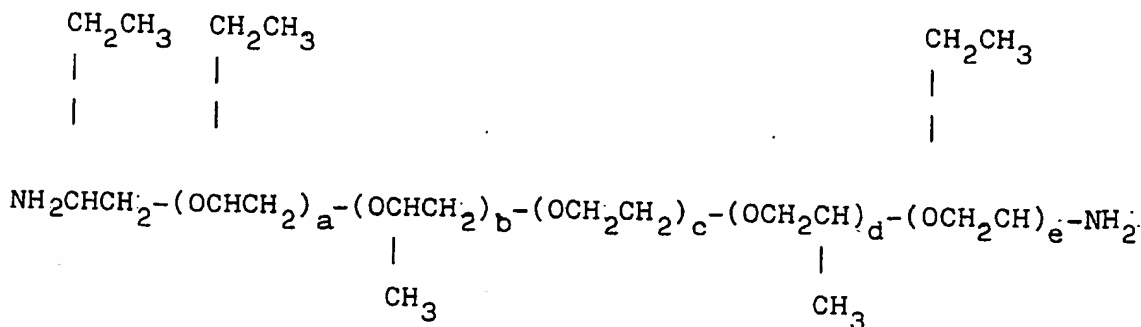


where R_4 is an aliphatic hydrocarbon radical having from 1 to 24 carbon atoms and n has a value of from 1 to 6.

- 25 2. A composition according to claim 1, in which said reaction product is obtained by reacting 1 mole of said dibasic acid anhydride with 1.5 moles of said polyoxyalkylene diamine and 1 mole of said hydrocarbyl polyamine.

3. A composition according to Claims 1 or 2, wherein said dibasic acid anhydride reactant is maleic anhydride.

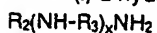
- 30 4. A composition according to Claims 1, 2 or 3, wherein said polyoxyalkylene diamine reactant is of the formula



where c has a value of from 8 to 50, b + d has a value of from 8 to 50, and a + e has a value of from 4 to 8.

- 45 5. A composition according to any one of the preceding claims, wherein said hydrocarbyl polyamine reactant is either:

(i) a hydrocarbyl polyamine of the formula



where R_2 is an alkyl radical having from 12 to 20 carbon atoms, R_3 is an alkylene radical having from 1 to 6 carbon atoms, and x has a value of from 1 to 5;

50 or

(ii) a n-alkyl-alkylene diamine of the formula



where R_4 is an aliphatic hydrocarbon radical having from 12 to 20 carbon atoms, and n has a value of 3.

- 55 6. A composition according to Claim 5, where said n-alkyl-alkylene diamine reactant is n-tallow-1,3 diaminopropane.

7. A concentrate composition comprising from 1.0 to 75.0 weight percent of the reaction product as claimed in any one of Claims 1 to 6 in admixture with a hydrocarbon solvent.

8. The use of a composition as claimed in any one of claims 1 to 6, or a concentrate as claimed in claim 7, as an ORI inhibitor additive for motor fuel.

9. A motor fuel composition comprising a mixture of hydrocarbons boiling in the range from 32°C to 232°C (90°F -450°F) and from 0.0005-5.0 weight percent of the reaction product as claimed in any one of claims 1 to 6.

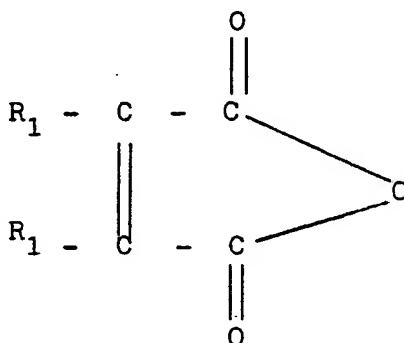
10. A motor fuel composition according to Claim 9 which additionally comprises from 0.001 to 1.0 weight percent of a polyolefin polymer, copolymer, or the corresponding hydrogenated polymer or copolymer, or mixtures thereof, of a C₂-C₆ unsaturated hydrocarbon, said polyolefin polymer or copolymer having a molecular weight in a range of from 500 to 3500.

11. A motor fuel composition according to Claim 10, in which said polyolefin polymer component is a polypropylene having a molecular weight in a range of from 750 to 1000.

12. A motor fuel composition according to Claim 10, in which said polyolefin polymer component is a polyisobutylene having a molecular weight in the range of from 1000 to 1500.

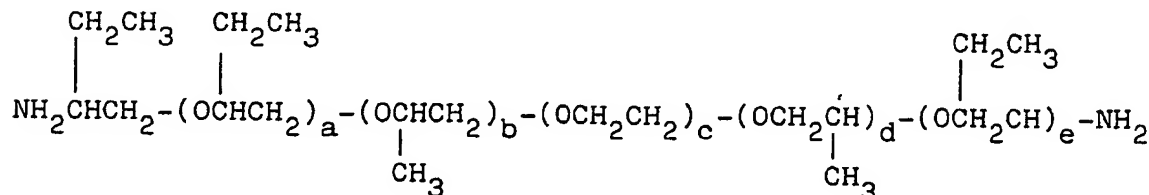
13. A method of manufacturing an ORI-inhibitor additive for motor fuel, characterised by the step of reacting at a temperature in a range of from 30°C to 200°C:

(a) about 1 mole of a dibasic acid anhydride of the formula



where R₁ is either H or a C₁-C₅ alkyl radical;

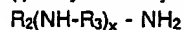
(b) 1-2 moles of a polyoxyalkylene diamine of the formula



where c has a value of from 5 to 150, b+d has a value of from 5 to 150, and a+e has a value of from 2 to 12; and

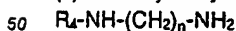
(c) 1-2 moles of a hydrocarbyl polyamine which may be either:

(i) a hydrocarbyl polyamine of the formula



where R₂ is a alkyl radical having from 1 to 24 carbon atoms, R₃ is an alkylene radical having from 1 to 6 carbon atoms, and x has a value of from 1 to 10; or

(ii) a n-alkyl-alkylene diamine of a formula



where R₄ is an aliphatic hydrocarbon radical having from 1 to 24 carbon atoms and n has a value of from 1 to 6.

FIG.1.

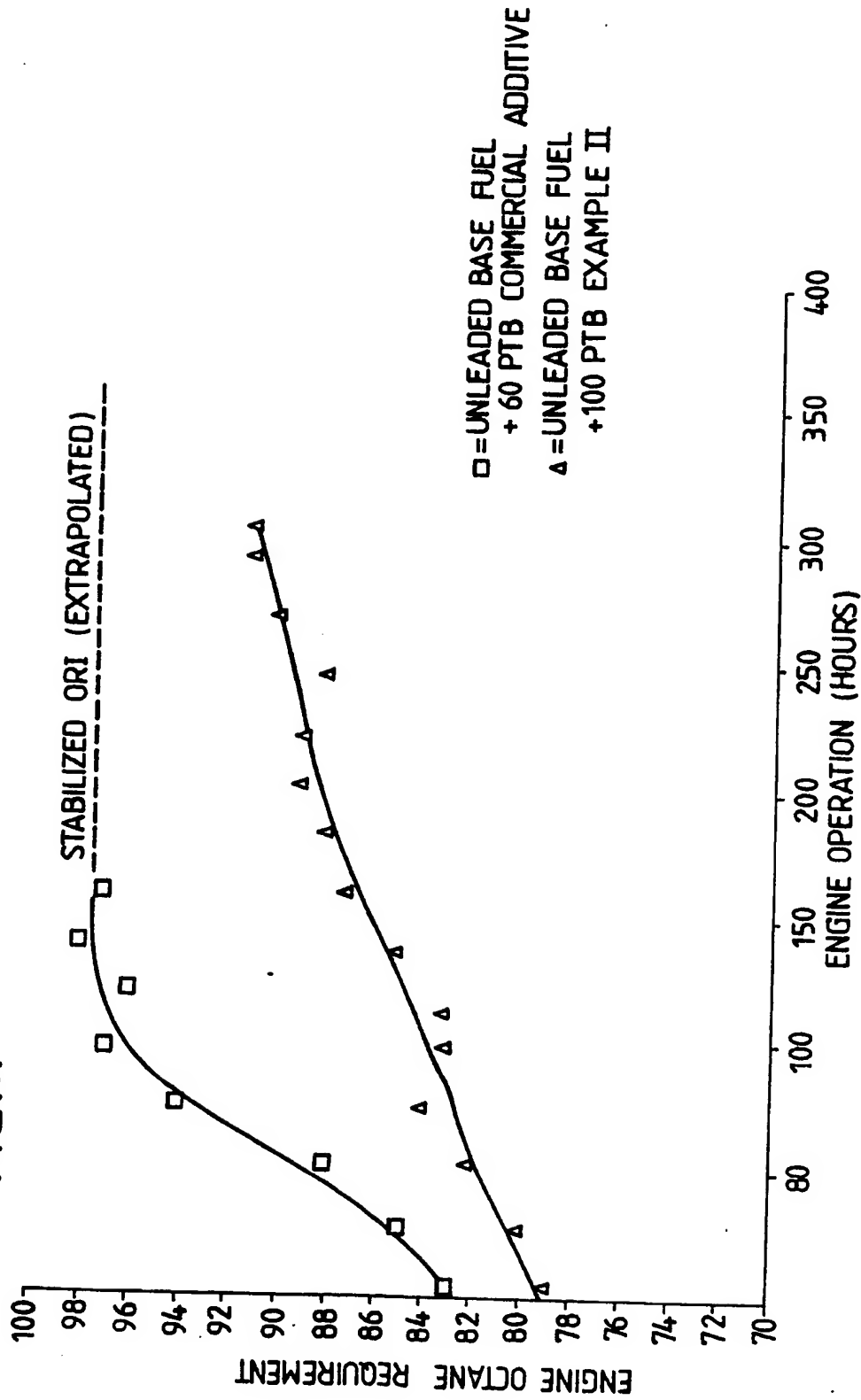
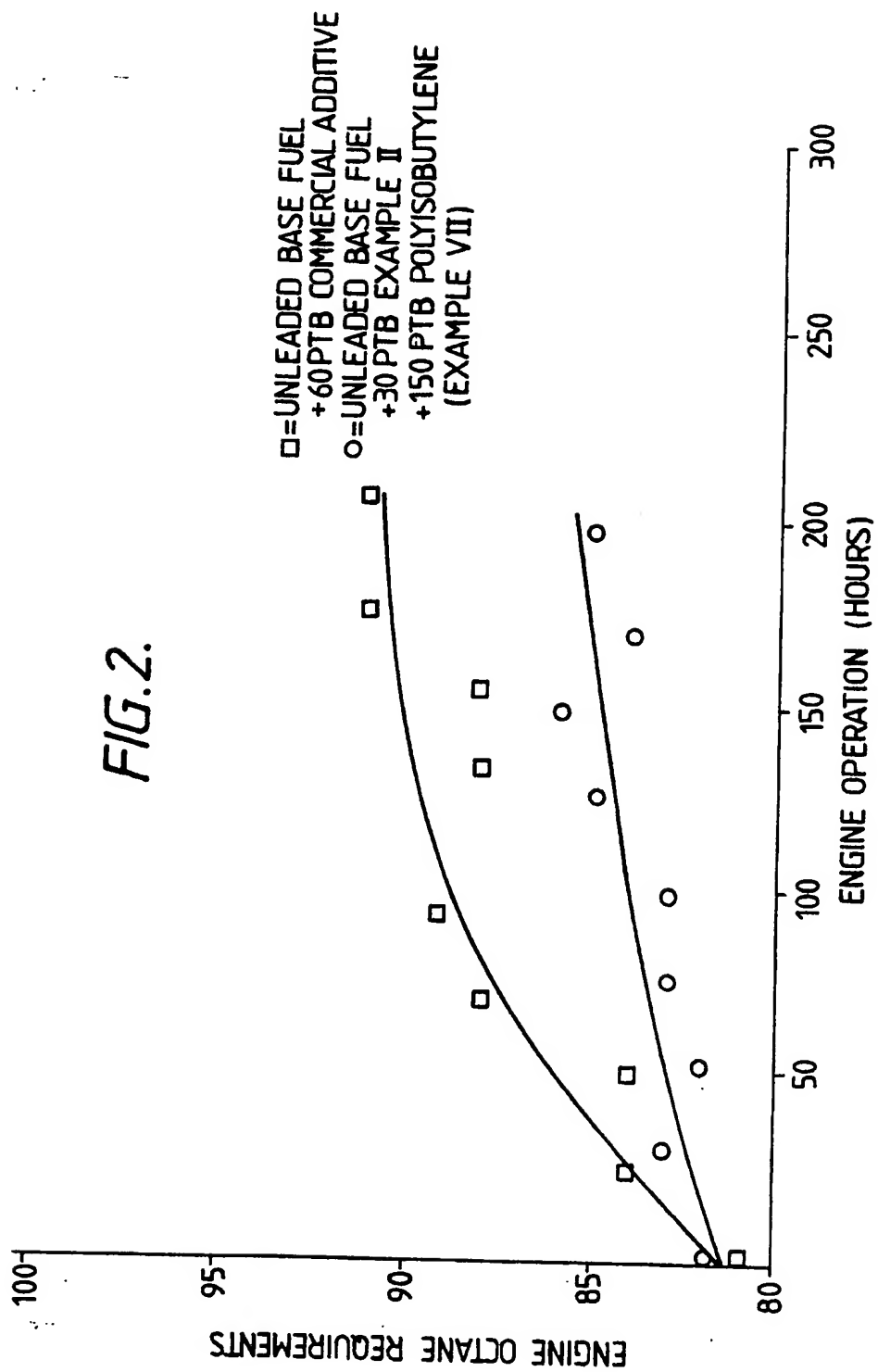


FIG. 2.





European Patent
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EUROPEAN SEARCH REPORT

Application Number

EP 87 30 8488

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.4)
A	US-A-4 631 069 (SUNG) * The whole document * ---	1-12	C 08 G 65/32 C 10 L 1/22 C 10 L 1/14
A	US-A-4 581 040 (SUNG et al.) * The whole document * ---	1-12	
A	US-A-4 107 061 (STURWOLD et al.) * The whole document * ---	1-12	
A	GB-A-1 173 788 (TEXACO) * Claims; page 4; page 2, lines 13-15 * ---	10-12	
A	US-A-4 181 682 (WATTS Jr. et al.) * The whole document * ---	1,4	
A	US-A-4 313 764 (TRACY et al.) * Column 2, line 16; column 3, lines 25-35 * ---	1,4	
A	GB-A-2 175 910 (TEXACO) * Claims * -----	1,4	
			TECHNICAL FIELDS SEARCHED (Int. Cl.4)
			C 08 G C 10 L
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 10-12-1987	Examiner DE LA MORINERIE B.M.S.B.
CATEGORY OF CITED DOCUMENTS			
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